Oxygen transport in *c*-axis-oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films, studied *in situ* by ellipsometry

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In this report we present a study on oxygen transport in *c*-axis-oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films. We have performed oxygen outdiffusion experiments, by gently heating these films in an ultrahigh-vacuum system. A rotating-analyzer ellipsometer has been used as an *in situ* monitor. For the oxygen transport we have developed an optical model. Our *in situ* ellipsometric measurements cannot be described without the assumption that short circuits are present in the films. The oxygen diffuses in the *a-b* planes of the $YBa_2Cu_3O_{7-\delta}$ to the short circuits. Along these short circuits, the oxygen leaves the films. In our case, the average spacing between the short circuits is 2.5 ± 0.2 μm. No outward diffusion of oxygen occurs along the *c* axis in the $YBa_2Cu_3O_{7-\delta}$ films.

I. INTRODUCTION

The properties of the high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ strongly depend on the oxygen stoichiometry. To obtain the highest critical superconducting transition temperature T_c, the oxygen deficiency δ should be very close to zero.[1] In most fabrication processes of high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films, a cooling down procedure is included in which the $YBa_2Cu_3O_{7-\delta}$ is transformed from an insulating tetragonal to the superconducting orthorhombic structure. During this treatment, δ changes from ≈1.0 to ≈0.0. To optimize the properties of $YBa_2Cu_3O_{7-\delta}$ thin films, the study of oxygen transport in these thin films is very important. For this purpose, spectroscopic ellipsometry (a nondestructive optical technique) is a very useful *in situ* monitor, yielding valuable information about the kinetics of the oxygen transport.

With ellipsometry, the ratio $\tilde{\rho}=\tan\Psi\exp(i\Delta)$ of the complex reflectances $\tilde{r}_p$ and $\tilde{r}_s$ of a sample is determined.[2] The indices *p* and *s* indicate the directions parallel (*p*) and normal (*s*) to the plane of incidence, defined by the photon paths and the normal to the sample surface, respectively. The complex reflectance ratio $\tilde{\rho}$ contains information about the complex dielectric function $\tilde{\epsilon}$ of materials and, therefore, about composition, microstructure, and thickness of different layers in the sample.

In the case of Cu-O-based ceramic high-T_c superconducting materials, spectroscopic ellipsometry has been used to determine $\tilde{\epsilon}$ of sintered polycrystalline superconducting $YBa_2Cu_3O_{7-\delta}$ samples and $YBa_2Cu_3O_{7-\delta}$ single crystals.[3,4] The effect of δ on $\tilde{\epsilon}$ has been investigated[4-6] and measurements have been compared with results on related Cu-O based materials.[7] The effects of metallic overlayers[8] and ionic substitutions for Y in $YBa_2Cu_3O_{7-\delta}$ (Ref. 9) have been studied. The effect of the superconducting transition of $YBa_2Cu_3O_{7-\delta}$ on $\tilde{\epsilon}$ has been determined.[10] Also the effects of the high anisotropy of $YBa_2Cu_3O_{7-\delta}$ on $\tilde{\epsilon}$ have been studied on $YBa_2Cu_3O_{7-\delta}$ single crystals and on *a*- and *c*-axes oriented thin films for $\delta=0.0$ (Refs. 11 and 12) and for $0.0\leq\delta\leq1.0$.[13] Synchrotron radiation has been used for spectroscopic ellipsometry on sintered polycrystalline superconducting $YBa_2Cu_3O_{7-\delta}$ samples.[14]

Here we present a study on the oxygen transport in $YBa_2Cu_3O_{7-\delta}$ thin films, monitored *in situ* with ellipsometry. Since the complex dielectric function of $YBa_2Cu_3O_{7-\delta}$ strongly depends on oxygen deficiency near photon energies of 4.0 eV,[4,5,13-15] ellipsometry is very well suited to detect oxygen deficiency and to monitor the oxygen transport *in situ*. For a more detailed analysis we have developed an optical model that enables us to determine the diffusion paths of oxygen through the *c*-axis oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films.

II. DIFFUSION IN A THIN FILM ON A SUBSTRATE

The diffusing species in a thin film on a substrate may enter or leave the film in two ways: through the solid-vacuum interface or along so-called short circuits, which may be present when the film is not crystalline. In Fig. 1 the film is assumed to be made up by square boxes, representing material that contains short circuits with an average spacing *s* in the *x* and *y* directions, respectively. The film thickness is denoted by *d*. For example, if in heteroepitaxial thin films grain boundaries act as source or drain for the diffusor, *s* equals the average spacing between these boundaries.

For the concentration *c* of the diffusing species, we may write[16]

$$\frac{\partial c}{\partial t}=-D_x(t)\frac{\partial^2 c}{\partial x^2}-D_y(t)\frac{\partial^2 c}{\partial y^2}-D_z(t)\frac{\partial^2 c}{\partial z^2}\ ,$$

with

$$D_i(t)=D_{i0}\exp\left[\frac{-w}{kT(t)}\right]\ ,\quad D_{i0}=\frac{r_i^2\nu}{q_i}\ ,i=x,y,z\ . \tag{1}$$

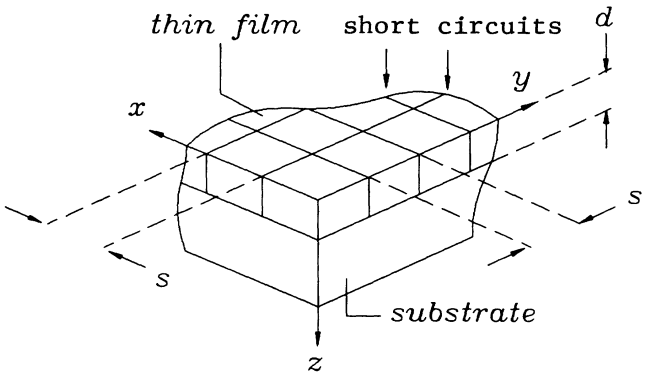

FIG. 1. Model for diffusion in a thin film on a substrate. The material in the film is separated by short circuits with an average spacing s. The walls of the boxes represent short circuits. For diffusion, two paths are available. The diffusor may diffuse to the film surface and desorb there. Also it is possible that the diffusing species migrates to a short circuit and leaves the film there. The film thickness equals d.

In this approach the diffusion coefficients D_{i0} are assumed to be only dependent on time t, not on c. However, D_{i0} may be different in the x, y, and z directions. The hopping activation energy equals w,, k is Boltzmann's constant. The temperature T may be time dependent. D_{i0} may be related to microscopic quantities[17]. The lattice constant of the material along the i axis is given by r_i, ν is the mean jump frequency, and q_i is the number of neighboring sites. We assume that no mass transport occurs at the substrate-film interface. We also assume that the diffusion process along the short circuits is a very fast process.

In the following we will develop three models for a diffusor leaving a thin film, i.e., due to a heat treatment. The boundary conditions describing the initial concentration profile c in the three different cases are the same:

$$c=c_0 \text{ for } 0<x<s\ , \quad 0<y<s\ , \quad 0<z<d\ , \text{ at } t=0\ . \tag{2}$$

Depending on the boundary conditions we apply for $t>0$ we can distinguish three different cases, which we call the 1D, 2D, and 3D diffusion models, respectively.

The 1D diffusion model describes the case in which the film contains very few or no short circuits. The average spacing s between short circuits then becomes very large. Diffusion along the x and y directions can be neglected, only diffusion along the z direction takes place. The desorption is assumed to be a very fast process. The boundary conditions for $t>0$ are

$$\frac{\partial c}{\partial x}=\frac{\partial c}{\partial y}=0 \text{ for } 0\le x\le s\ ,\ 0\le y\le s\ ,\ 0\le z\le d\ ,$$
$$c=0 \text{ for } z=0\ ,\ 0<x<s\ ,\ 0<y<s, \text{ and } \frac{\partial c}{\partial z}=0 \text{ for } z=d\ . \tag{3a}$$

The solution for the 1D diffusion model is given by[16]

$$c(z,t)=c_0 f(z,2d,D_z)\ , \tag{3b}$$

where

$$f(p,q,D_i)=\frac{4}{\pi}\sum_{j=0}^{\infty}\frac{1}{2j+1}\sin\left[\frac{(2j+1)\pi p}{q}\right]\exp\left[-\left[\frac{(2j+1)\pi}{q}\right]^2 D_i t\right]\ . \tag{3c}$$

If the film-vacuum interface acts as a diffusion barrier or if D_z is negligibly small, we obtain the 2D diffusion model. No diffusion along the z axis occurs. The diffusor only migrates along the x and y directions and leaves the film via short circuits. The boundary conditions for $t>0$ are

$$c=0 \text{ for } x=0 \text{ and } x=s\ ,\ 0<y<s\ ,\ 0<z<d\ ,$$
$$\text{for } y=0 \text{ and } y=s\ ,\ 0<x<s\ \ 0<z<d, \text{ and} \tag{4a}$$
$$\frac{\partial c}{\partial z}=0 \text{ for } 0\le x\le s\ ,\ 0\le y\le s\ ,\ 0\le z\le d\ .$$

The solution for the 2D diffusion model is given by

$$c(x,y,t)=c_0 f(x,s,D_x)f(y,s,D_y)\ . \tag{4b}$$

In the 3D diffusion model we assume that the desorption from the film surface is a very fast process. The outdiffusion process is diffusion limited. The corresponding boundary conditions for $t>0$ are

$$c=0 \text{ for } x=0 \text{ and } x=s\ ,\ 0<y<s\ ,\ 0<z<d\ ,$$
$$\text{for } y=0 \text{ and } y=s\ ,\ 0<x<s\ ,\ 0<z<d\ , \tag{5a}$$
$$\text{for } z=0,\ 0<x<s\ ,\ 0<y<s\ , \text{ and}$$
$$\frac{\partial c}{\partial z}=0 \text{ for } z=d\ .$$

The solution equals

$$c(x,y,z,t)=c_0 f(x,s,D_x)f(y,s,D_y)f(z,2d,D_z)\ . \quad (5b)$$

This solution describes the case, in which diffusion in all three directions plays a role.

The quantity $\overline{D_i t}$ in Eq. (3c) is defined as

$$\overline{D_i t}=\int_0^t D_i(t)dt\ . \quad (6)$$

When $T(t)$ is measured, $\overline{D_i t}$ can be calculated numerically using Eq. (1) with values for D_{i0} and w taken from literature.

III. OPTICAL MODEL FOR OXYGEN OUTDIFFUSION EXPERIMENTS ON $YBa_2Cu_3O_{7-\delta}$ THIN FILMS, MONITORED *in situ* BY ELLIPSOMETRY

When a high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ ($\delta\approx 0.0$) thin film is subject to a gentle heat treatment at a very low oxygen background pressure, oxygen will diffuse out of the thin film. For $0.0<\delta<1.0$, the oxygen at the O(1) sites is the diffusing species.[18–20] The fraction of vacant O(1) sites equals δ. So c_0 in Eq. (2) is equal to N_0, the concentration of O(1) sites (1 per unit cell).

It is known that in randomly twinned $YBa_2Cu_3O_{7-\delta}$ the order of magnitude of the diffusion coefficients along the a and b axes are comparable, whereas the diffusion coefficient along the c axis is much smaller.[19,20] Since we performed experiments on c-axis oriented $YBa_2Cu_3O_{7-\delta}$ films and twinning cannot be excluded, D_{x0} and D_{y0} were chosen to equal 1.4×10^{-4} cm^2/s. D_{z0} may be approximated by 0.001 $D_{x,y0}$. For w a value of 0.97 eV can be found in the literature.[19,20]

The three diffusion models described by Eqs. (3)–(5), yield us c as a function of position and time. Since we cannot measure c directly by ellipsometry, we have to relate c to the complex dielectric function $\tilde{\epsilon}_{Y-Ba-Cu-O}=\epsilon_1-j\epsilon_2$ of $YBa_2Cu_3O_{7-\delta}$. We performed *in situ* measurements at three photon energies: 4.0, 4.1, and 4.2 eV. In this report results of measurements at 4.0 eV are shown. Experiments at the two other energies gave similar results. With the phenomenological equation (using data from Refs. 13 and 15)

$$\epsilon_1=2.5+3.8\delta\ ,\quad \epsilon_2=2.0-3.2\delta+9.1\delta^2\ , \quad (7)$$

and $\delta=1-c(x,y,z,t)/N_0$, we can calculate $\tilde{\epsilon}_{Y-Ba-Cu-O}(x,y,z,t)$ at a photon energy of 4.0 eV (at room temperature). Knowing $\tilde{\epsilon}_{Y-Ba-Cu-O}(x,y,z,t)$, we may use an optical multilayer model[2] for the 1D diffusion model. In the case of 2D and 3D diffusions, the multilayer model may be combined with a Lorentz-Lorentz effective medium approximation[21] to calculate theoretical values for Δ and Ψ. These values can then be compared with the experimental data. From this comparison we can conclude which model (1D, 2D, or 3D diffusion, respectively) describes the experiments best.

The most relevant parameters in our model are (I) the diffusion coefficients D_{i0} and hopping activation energy w, (II) the average spacing s between the short circuits, and (III) the film thickness d [see Eqs. (3)–(5)]. The film thickness can be determined by ellipsometric measurements, as we showed before.[15] Values for D_{i0} and w may be taken from the literature.[19,20] The mean spacing s between short circuits is the only adjustable parameter in our optical model.

IV. EXPERIMENTAL

High-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films were deposited on yttria-stabilized ZrO_2 (*YSZ*) (100) single crystals using a modified off-axis rf-magnetron sputtering technique.[22] X-ray diffraction analysis (XRD) was used to determine the structure and orientation of the $YBa_2Cu_3O_{7-\delta}$ layers. The length of the c axis of the different $YBa_2Cu_3O_{7-\delta}$ films was determined using XRD. Superconducting properties were derived from measurements of the critical temperature T_c and the critical current density j_c.

A Debye-Scherrer diffractometer with a Cu $K\alpha$ x-ray source was used for XRD analysis. Ellipsometry was performed using a fully computer controlled rotating-analyzer ellipsometer (RAE), already described elsewhere.[23–25] The RAE is mounted on two optical benches, which are attached to an ultrahigh vacuum (UHV) system. The UHV system, with a base pressure of 2×10^{-10} mbar, is equipped with a mass spectrometer for residual gas analysis. Inside the UHV system, the [YSZ(100)]/$YBa_2Cu_3O_{7-\delta}$ samples are mounted on a Si substrate. The Si substrate can be resistively heated by a computer controlled current source. In this way the $YBa_2Cu_3O_{7-\delta}$ thin films can be given a controllable heat treatment. A thermocouple is mounted on the Si substrate, allowing *in situ* determination of the Si substrate temperature.

On a set of samples oxygen outdiffusion experiments have been done. First the layer thicknesses were determined with ellipsometry.[15] Oxygen transport in c-axis oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films has been investigated by carefully heating the $YBa_2Cu_3O_{7-\delta}$ films in the UHV system, causing oxygen to diffuse out of the samples. The heat treatment has been monitored *in situ* with the RAE. Δ and Ψ have been measured at incident photon energies of 4.0, 4.1, and 4.2 eV during the warm up of the samples, the oxygen outdiffusion, and the cooling down of the $YBa_2Cu_3O_{7-\delta}$ thin films. Simultaneously to the RAE measurements, the temperature of the thermocouple has been measured. The length of the c axis in $YBa_2Cu_3O_{7-\delta}$ is related to the value of δ.[18] With XRD, the length of the c axis in $YBa_2Cu_3O_{7-\delta}$ thin film can be determined and, therefore, an estimate of δ (averaged of the detection volume) after the gentle heat treatment can be obtained.

V. RESULTS

By means of the modified rf-magnetron sputtering technique, as mentioned in Sec. IV, high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films with a transition temperature $T_{c,zero}$ of about 88 K were routinely obtained for film thicknesses of 8–300 nm. The critical current densi-

ty j_c at 77 K of these films is found to be higher than 1×10^6 A/cm^2. With XRD, besides the substrate reflections only the (00l) reflections could be observed.[26]

Experiments have been performed on a set of samples. In this paper typical results on one of these samples are presented. Other experiments showed similar results. By ellipsometry, the film thickness of sample 1 was determined to be 67±7 nm. Before the heat treatment, its $T_{c,\mathrm{zero}}$ equaled 85.7 K. Results of *in situ* ellipsometric measurements on the heat treatment of sample 1 are given in Fig. 2. The measurements have been performed at a photon energy of 4.0 eV. At $t=0.2$ h, the heat treatment was started. Above $T=470$ K (reached at $t=0.3$ h), the first effect of oxygen diffusion out of the $YBa_2Cu_3O_{7-\delta}$ thin film can be observed. Δ and Ψ start to change. At that point (by means of a mass spectrometer tuned to the O_2 mass), the O_2 partial pressure in the UHV chamber was observed to increase from 2×10^{-11} to 4×10^{-10} mbar, indicating oxygen outdiffusion. It should be noted that during the initial heating of the sample ($t<0.3$ h) no temperature effect on Δ and Ψ can be observed. If the temperature is further increased, oxygen diffuses out and the oxygen concentration in the film decreases further, as can be seen from the changes in Δ and Ψ. At $t=1.6$ h the heat treatment was stopped. In contrast to the initial heating run, a strong temperature effect on Δ and Ψ can be seen during the cool down of the sample, especially on Ψ. That this is indeed a temperature effect, has been confirmed by gently varying the temperature of the sample after it had cooled down. Δ and Ψ reversibly changed with temperature in the range 300–450 K. With XRD an estimate of the oxygen deficiency δ can be obtained, since the length of the c axis is related to the oxygen concentration in the $YBa_2Cu_3O_{7-\delta}$.[18] Due to the heat treatment, the length of the c axis of the $YBa_2Cu_3O_{7-\delta}$ thin film changed from 1.167±0.001 nm to 1.182±0.002 nm, corresponding to a change in δ from 0.0 to 0.9.[18] After this heat treatment, the $YBa_2Cu_3O_{7-\delta}$ thin film did not show a superconducting transition anymore.

In Fig. 3 the *in situ* ellipsometric measurements at a photon energy of 4.0 eV on the heat treatment of sample 1 are given as a trajectory in the (Δ,Ψ) plane, indicated by "meas." The heat treatment starts at the point indicated by "i." At the point indicated by the arrow "e" the heat treatment was stopped. The total heat treatment took about 2.4 h, see Fig. 2. The same strong temperature effect on Δ and Ψ, already shown in Fig. 2, can be seen in Fig. 3 during cool down of the sample. The trajectory runs from the point indicated by "e" to point "f." A first-order correction was performed on the data, the results are indicated by "corr."

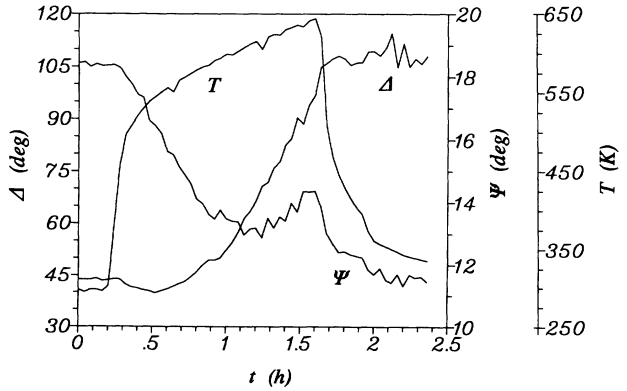

FIG. 2. Δ, Ψ, and T measured *in situ* as a function of time during the gentle heat treatment of an c-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin film (sample 1).

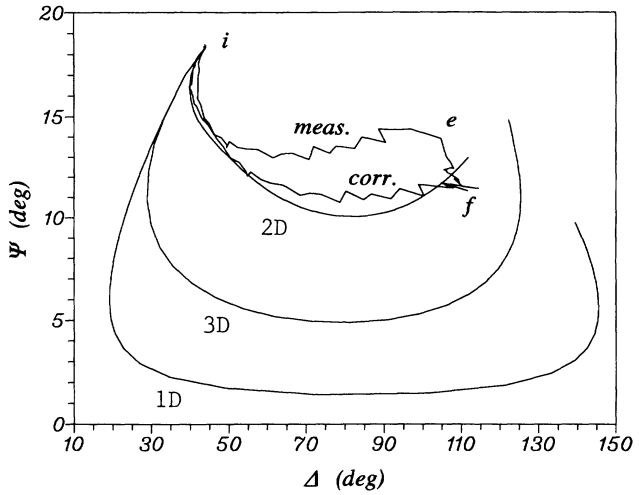

FIG. 3. *In situ* ellipsometric measurements at a photon energy of 4.0 eV on the heat treatment of an c-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin film (sample 1), indicated by "meas." The trajectory runs from "i" via "e" to "f." Note the temperature effect from "e" to "f" on Δ and Ψ when the sample is cooled down. A first-order correction has been performed and the results are indicated by "corr." For δ, averaged over the detection volume, a value of 0.9 after the heat treatment has been found. The results of the optical outdiffusion models are indicated by 3D, 2D, and 1D, respectively. For a description, see Secs. II, III, and V.

The theoretical results of the optical models we have developed in Secs. II and III are given as solid lines in Fig. 3 and are indicated by 1D, 2D, and 3D, respectively. In 1D diffusion, s plays no role, see Eq. (3b). What can be seen immediately, is that the measurements are very poorly described by a 1D diffusion process. In the 2D and 3D diffusion processes, s plays a very important role. The quantities $\overline{D_x t}$, $\overline{D_y t}$, and s are coupled in the solutions, see Eqs. (4b) and (5b). Therefore, adjusting s mainly affects the time at which the oxygen outdiffusion is completed. By increasing s, the oxygen has to diffuse over greater distances in the material to the short circuits. The effect will be that the decrease of δ, averaged over the detection volume, is slower. We have measured the temperature T as a function of time, so we can calculate $\overline{D_i t}$ [see Eqs. (1) and (6)]. s has been adjusted in order to match the end points of the theoretical and experimental data (the points indicated by "f"). We have found that s equals 2.5±0.2 μm and the results, indicated by 1D, 2D, and 3D, respectively, are shown in Fig. 3. As can be observed, also the 3D diffusion model poorly de-

scribes the measurements. The oxygen outdiffusion measurements can be described by a 2D diffusion process very well.

VI. DISCUSSION

The $YBa_2Cu_3O_{7-\delta}$ thin film is assumed to consist of *c*-axis oriented crystalline material, that contains short circuits with an average spacing of 2.5 ± 0.2 μm. We would like to discuss two possible features that may act as short circuits in $YBa_2Cu_3O_{7-\delta}$ thin films.

Scanning tunneling microscopy (STM) and transmission electron microscopy (TEM) studies reveal that $YBa_2Cu_3O_{7-\delta}$ thin films contain growth spirals that started at screw dislocations, present on the substrate surface.[27,28] Defects may occur when during film growth the growth spirals meet. At the spiral boundaries dislocation defects, point defects, and out-of-phase domains may be expected. These boundaries very well may act as short circuits. For the growth spiral density a value of about $10^{12}-10^{14}$ m^{-2} (Ref. 27) and 10^{13} m^{-2} (Ref. 28) has been found. The screw dislocation density strongly depends on growth temperature and substrate misorientation. Our results correspond to a screw dislocation density of 1.6×10^{11} m^{-2}, a relatively low value. Secondly, boundaries between the *c*-axis oriented $YBa_2Cu_3O_{7-\delta}$ material and outgrowths, which in general are present in the films, may act as short circuits. TEM studies indicate that these outgrowths are *a*-axis oriented $YBa_2Cu_3O_{7-\delta}$ grains.[29,30] Typical distances between outgrowths are in the range $1-10$ μm, again very dependent on preparation conditions. Also these values correspond well to our results. We may conclude that with our method we cannot determine what the short circuits exactly are, but the assumption of their presence is a necessary condition to describe the results of our *in situ* ellipsometric measurements on oxygen outdiffusion. So we may conclude that short circuits are present in our $YBa_2Cu_3O_{7-\delta}$ thin films which are active at least in the temperature range 450–650 K.

In our optical model we used values for D_{i0} and w from literature.[19,20] Since diffusion in $YBa_2Cu_3O_{7-\delta}$ is highly anisotropic, the orientation and microstructure of the samples in diffusion experiments are very important. Grain sizes, twinning, composition, and structure of grain boundaries may very well influence experimental results. Scatter of data may be expected, due to differing preparation techniques.[31–33] Surprisingly, deviation of these data does not affect our results very much. As is shown in Eq. (4b), the 2D diffusion case, $\overline{D_i t}$ and s are coupled. Uncertainties in $\overline{D_i t}$ only will show up as scattering in s. The shape of the theoretical trajectories in Fig. 3 remains nearly unchanged. So our observation that the oxygen outdiffusion in *c*-axis oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films is a 2D process, is not weakened by variations in diffusion coefficients and/or activation energies.

In our model we used a phenomenological equation to relate the complex dielectric constant of $YBa_2Cu_3O_{7-\delta}$ at 4.0 eV to the oxygen deficiency δ. Since this relation may be not accurately known yet, some deviation between experimental and theoretical data may be expected. Also the method to correct for the temperature effect in the complex dielectric constant may be a point for discussion. We used a simple first-order correction. However, the very good agreement between theory and experiment in the 2D diffusion model, indicates that Eq. (7) describes the dielectric constant of $YBa_2Cu_3O_{7-\delta}$ at 4.0 eV fairly well as a function of δ.

We assumed that $D_{c0}=10^{-3}D_{a,b0}$.[19] Also it is found that $D_{c0}=10^{-6}D_{a,b0}$.[20] In the first case we observe in Fig. 3 that the diffusion along the *c* axis may not be neglected. So in this case the surface of the thin film acts as a diffusion barrier, leading to a 2D diffusion model. Numerical calculations showed that in the second case the diffusion along the *c* axis may be neglected. D_{c0} is very small compared to $D_{a,b0}$, which also leads to a 2D diffusion model. Whether the very low D_{c0} is intrinsic to the $YBa_2Cu_3O_{7-\delta}$ material or due to a surface barrier is not fully clear yet. However, our results clearly show that during the oxygen outdiffusion experiments on our *c*-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin films, no outdiffusion occurs along the *c* axis.

In this paper, results of oxygen outdiffusion experiments on $YBa_2Cu_3O_{7-\delta}$ films are presented. No outdiffusion along the *c* axis takes place. If oxygen indiffusion is possible along the *c* axis, it has to be investigated. However, oxygen indiffusion along short circuits and, subsequently, along the *a* and *b* directions into the *c*-axis oriented $YBa_2Cu_3O_{7-\delta}$ material has to be expected. Our optical models easily can be adapted to describe *in situ* ellipsometric measurements on oxygen indiffusion experiments.

VII. CONCLUSIONS

We have found that oxygen outdiffusion in *c*-axis oriented high-T_c superconducting $YBa_2Cu_3O_{7-\delta}$ thin films is a 2D diffusion process. A rotating-analyzer ellipsometer (RAE) has been used as an *in situ* monitor. Our *in situ* ellipsometric measurements cannot be described without the assumption that in the films short circuits are present. The oxygen diffuses in the *a-b* planes of the $YBa_2Cu_3O_{7-\delta}$ to the short circuits. Along these short circuits the oxygen leaves the films. In our case the average spacing between the short circuits is 2.5 ± 0.2 μm. No oxygen outdiffusion occurs along the *c* axis in the $YBa_2Cu_3O_{7-\delta}$ films.

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